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A Technical Review of Non-Destructive Assay Research for the Characterization of Spent Nuclear Fuel Assemblies Being Conducted Under The US DOE NGSI

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Abstract

There is a growing belief that expansion of nuclear energy generation will be needed in the coming decades as part of a mixed supply chain to meet global energy demand. At stake is the health of the economic engine that delivers human prosperity. As a consequence renewed interest is being paid to the safe management of spent nuclear fuel (SNF) and the plutonium it contains. In addition to being an economically valuable resource because it can be used to construct explosive devices, Pu must be placed on an inventory and handled securely. A multi-institutional team of diverse specialists has been assembled under a project funded by the US Department of Energy (DOE) Next Generation Safeguards Initiative (NGSI) to address ways to nondestructively quantify the plutonium content of spent nuclear fuel assemblies, and to also detect the potential diversion of pins from those assemblies. Studies are underway using mostly Monte Carlo tools to assess the feasibility, individual and collective performance capability of some fourteen nondestructive assay methods. Some of the methods are familiar but are being applied in a new way against a challenging target which is being represented with a higher degree of realism in simulation space than has been done before, while other methods are novel.

In this work we provide a brief review of the techniques being studied and highlight the main achievements to date. We also draw attention to the deficiencies identified in for example modeling capability and available basic nuclear data. We conclude that this is an exciting time to be working in the NDA field and that much work, both fundamental and applied, remains ahead if we are to advance the state of the practice to meet the challenges posed to domestic and international safeguards by the expansion of nuclear energy together with the emergence of alternative fuel cycles.

Introduction

Energy security and how to achieve it in a sustainable and environmentally responsible way is a major global challenge. Many experts are of the opinion that nuclear energy is likely to not just remain an important part of the energy supply mix but to expand over the coming decades. It seems that the next generation of commercial nuclear power plants will continue to be predominantly variants of light water reactor designs. For many years the United States has operated a sizable fleet of light water reactors (~104) according to a once through fuel cycle. Spent nuclear fuel (SNF) assemblies are stored at reactor sites. Currently there is no deep geologic nuclear repository or other long term management option. Indeed by 2020 it is expected that the US will have more SNF than the Yucca Mountain repository proposed in the 1970's, but currently without a timeline for being completed, could accommodate. Thus the stewardship of SNF is a high priority. Most of the plutonium in the world is present in commercial spent fuel and is therefore in need of responsible safeguarding to prevent the misuse of this potential nuclear explosive.

The Next Generation Safeguards Initiative (NGSI) of the U.S. Department of Energy (DOE) is supporting a multi-laboratory / university collaboration to assess ways to quantify plutonium mass in SNF assemblies and to also detect the diversion of pins for such assemblies by non-destructive assay (NDA) methods. Fourteen promising techniques were identified for evaluation. A phased program of work was begun with the initial focus on modeling studies to establish feasibility and capability of each method implemented as an independent prototypical instrument and to determine how best to integrate some sub-set of techniques into a comprehensive NDA system offering an enhanced capability over present state of the practice for commercial light water reactor fuels. Some of the techniques are familiar although they may not have been applied to SNF assembly measurements in the past. Others are just emerging so that the level of understanding is less developed and formidable technical challenges must be overcome before they can be considered mature and ready for use. The 14 NDA techniques which we shall review here are:

DDA - Differential Dieaway
DN – Delayed Neutron
DG – Delayed Gamma
PG – Passive prompt Gamma spectroscopy
PNAR-3He and PNAR-FC – Passive Neutron Albedo Reactivity with 3He proportional counters of fission chambers
DDSI – Differential Dieaway Self-Interrogation
GN – Gross (or Total) Neutron
NM – Neutron Multiplicity
CIPN – ^{252}Cf Interrogation with Prompt Neutron Detection

SINRD – Self-Interrogation Neutron Resonance Densitometry
XRF – X-Ray Fluorescence
LSDS – Lead Slowing Down-time Spectrometry
NRTA – Nuclear Resonance Transmission Analysis
NRF – Nuclear Resonance Fluorescence

For each technique we shall highlight the main project achievements to date and flag the immediate challenges. In order to evaluate the performance the instruments have been pitted against a synthetic spent fuel library covering the range of commercial pressurized water reactor fuel conditions expected [Fensin et al 2009].

An Integrated Delayed Neutron and Differential Die-Away Instrument with ^{3}He Detectors and a DT Generator

The neutronic design of the DN instrument and the DDA are similar and so integrate natural in simulation space. Here we use a ^{3}He proportional counter based detector system and a sealed DT neutron generator. DN counting is an active assay technique that consists of turning on and off the interrogating DT source and counting the number of delayed neutrons emitted when the source is off. First a passive neutron measurement is performed to determine the background count rate (singles), mainly originating from spontaneous fission of ^{244}Cm in spent fuel and amplified by self-multiplication. An active assay is then undertaken by switching on the DT generator. Spectrum tailoring was implemented to lower the energy of the ~14.1 MeV neutrons created before they enter the fuel. The time dependence of the delayed neutrons emitted from the many precursors produced is described by only six groups[1] in the code used for all simulations (MCNPX 27c). The effective half-lives of these groups vary from ~0.2 sec to ~1 minute. The safeguards goal of interest to this work is quantifying the mass of elemental Pu and thus the fissile content mainly dominated by delayed neutrons from ^{235}U, ^{239}Pu and ^{241}Pu, prominent fissile isotopes of interest. However the fission of the main fertile, ^{238}U can be a significant issue since it has the largest mass of any isotope in the fuel. The delayed neutron instrument was designed to emphasize the signal from the fissile isotopes relative to fertile isotopes. Through later integration with other NDA instruments, the fissile Pu will be used to determine elemental Pu. The Differential Die-Away (DDA) measure prompt induced fission neutrons (PN), could allow us to get the PN/DN ratio to obtain the mass of Pu. The instrument is showed in Figure 1 below.

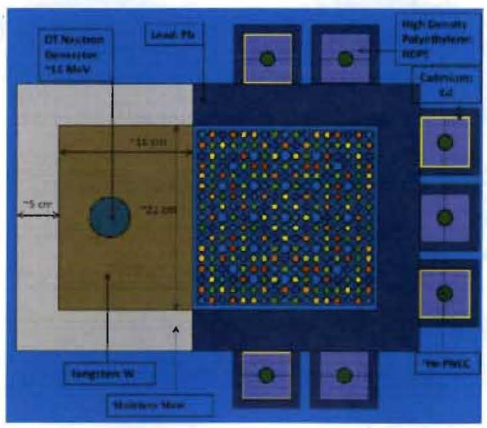

Figure 1: XY slice of the design of the delayed neutron instrument using a DT generator and ^{3}He detectors.

The main results of the study lead on the delayed neutron instruments are the following, (1) For 45 GWd/tU burnt assemblies, removing 40 pins from any region is easily detectable as well as removing only 24 pins from the inner region of the 17 x 17 pin array. (2) According to the

Signal to Background Ratio (S/B) in the context of this design using a DT Generator and ^{3}He detectors in order to achieve a S/B of ~30% throughout the 64 FA of the spent fuel library a source strength 5 x 10^{10} n/s to 10^{11} n/s should be used. (3) The weight of each detectors to DN detection has been determined and could allow to optimize the design when integration with another NDA instrument. (4) The fissile content has been determined via the $^{239}Pu_{eff\text{-}DN}$ mass as observed in Figure 2. (5) Other designs have been explored and show that in water the design doesn't impact tremendously the results and they happen to be very similar on almost every aspect and (6) Other media have not been explored since the DN is aimed to be integrated to the DDA which isn't expected to be usable in either borated water or air.

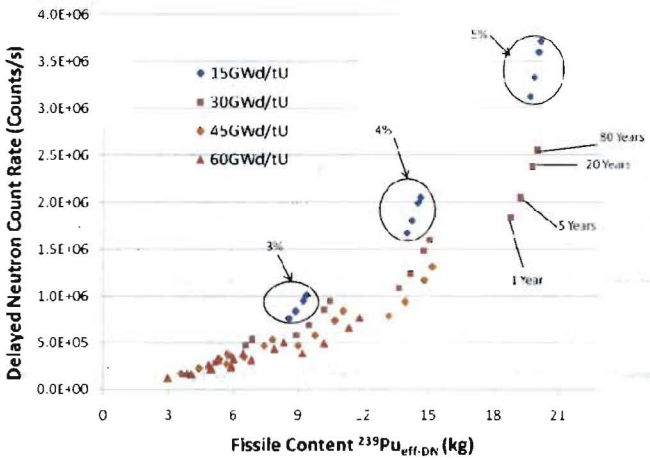

Figure 2: Delayed neutron count rate as a function of the fissile content / mass of $^{239}Pu_{eff\text{-}DN}$ for the 64 fuel assemblies of the spent fuel library representing 4 burnups, 15, 30, 45 and 60 GWd/tU, 4 cooling-times, 1, 5, 20 and 80 years and 4 initial enrichments.

C.J. Werner "Simulation of Delayed Neutrons Using MCNP," Progress In Nuclear Energy, Vol. 41, PJI: S0149-1970(02)00019-7, 2002 Elsevier Science.

Differential Dieaway System

The same detector design has been used to study the performance of a differential dieaway (DDA) instrument, although a different source tailoring scheme was used. The DDA technique is also an active neutron assay technique and has been used to assay the fissile content of radioactive waste drums [2] and cargos [3]. The application of DDA technique to assay spent fuel assemblies (SFA) is a new effort. The interrogating source neutrons are thermalized in the water pool of the spent fuel by the water medium, and create fission reactions in the fissile material of spent fuel assemblies. The neutrons from the generator cause fission reactions in the fuel that produce a new source of fast neutrons that is significant because the multiplication is greater than two. The only detected neutrons in Cd-covered ^{3}He tubes are these fast neutrons and the population of these induced fission neutrons decays with a die-away time of several hundred microseconds because of the multiplication chains. This die-away time varies according to the contents of fissile isotopes such as ^{235}U, ^{239}Pu, and ^{241}Pu contained in the spent fuel assembly. With more fissile content, the die- away time gets longer. The neutron die-away time of a pure $^{238}UO_2$ assembly (i.e. no fissile material) is much shorter than those of the other SFAs. This is because the sustaining time of chain reaction of ^{238}U isotope by the thermal neutron is much shorter than those of fissile isotopes.

The MCNPX [4] simulations of DDA technique were performed for 64 cases of the spent fuel library in water. We have demonstrated that the counting rate in the differential dieaway window (0.2-1ms) is a simple function of $^{239}Pu_{effective}$ for simplified assemblies composed of uranium and plutonium oxide. However for the spent fuel library set of 64 assemblies, the counting rate becomes a relatively complex function of $^{239}Pu_{effective}$ because of the buildup of neutron absorbers, the amounts of which depend on burnup, initial enrichment and cooling time. The counting rate depends on the effective multiplication of the assembly. This multiplication has a reasonably simple relationship with the observed dieaway time of the measurement. As with other techniques the plutonium mass in the fuel cannot be obtained directly from this method. One option would be to establish calibration curves for different burnup and cooling time and these parameters would be obtained from other measurement techniques.

For neutron signal measurements of a spent fuel assembly, there exists a very intense neutron background coming mainly from spontaneous fission of ^{244}Cm. If we ignore some unrealistic cases such as 2% IE, 45 and 60 GWD/MTU and 3% IE and 60 GWD/MTU, we can obtain the signal to background ratio of at least 5 for the remaining 52 cases with the neutron generator of 1 $\times 10^{9}$ n/s strength. This strength of neutron generator is readily available commercially.

The study only focused on the application of DDA technique for the spent fuel measurement in water. Additional studies for borated water or air applications of DDA technique are needed.

1. Tae-Hoon Lee, H. O. Menlove, M. T. Swinhoe, S. J. Tobin "Differential Die-Away Technique for Determination of the Fissile Contents in Spent Fuel Assembly," Proc 51st INMM Annual meeting Baltimore USA (2010).

2. J. T. Caldwell, R. D. Hastings, G. C. Herrera and W. E. Kunz, et al., "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum Size Containers," Los Alamos National Laboratory Report, LA-10774-MS (1986).

3. K. A. Jordan and T. Gozani, "Pulsed Neutron Differential Die Away Analysis for Detection of Nuclear Materials," Nucl. Instruments Methods Phys. Res., vol B 261, pp. 365-368 (2007).

4. D. B. Pelowitz et al., "MCNPX User's Manual, Version 2.7.B Extensions," Los Alamos National Laboratory Report, LA-UR-09-04 150 (2009).

Delayed Gamma

In the context of the NGSI project, a delayed gamma non-destructive assay (NDA) technique is being investigated as (1) a means to directly quantify both the fissile and fertile content of the spent nuclear fuel, and (2) as a general safeguards tool that can be easily integrated with other active interrogation instruments.

In support of this research, a newly developed photon assay modeling approach was introduced. It integrates modified versions of existing Monte Carlo-based transport (MCNPX) and analytical decay/depletion (CINDER) codes with a specifically-written Discrete Gamma Source DEFinition (DGSDEF) code. The resulting hybrid simulation scheme provides robust calculations for time- and spatially-dependent passive and actively induced discrete gamma source terms and detector responses [2]. The performance of this calculation technique was benchmarked and validated in the extensive experimental campaign at the Idaho Accelerator Center involving accelerator-driven neutron sources and samples of fissile and fertile materials and their combinations with varying parameters of interrogation setups. Data from other experimental sites was also incorporated. Following the good benchmark performance, the modeling approach and the DGSDEF code as a stand-alone application were copyrighted and approved for a limited release at the Los Alamos National Laboratory [3]. Currently, the work is geared at merging the DGSDEF capability into the new version of the CINDER code for subsequent public release. As a result of the code development effort, application of the new modeling technique was extended to the design process of four photon-based NDA techniques considered under the NGSI project: (1) delayed gamma, (2) x-ray fluorescence, (3) nuclear resonance fluorescence, and (4) passive gamma – plutonium isotopic correlation.

The capability of a delayed gamma spent nuclear fuel assay system was analyzed both for a stand-alone configuration and in integration with delayed neutron and differential die-away techniques also considered under the NGSI effort. Variations and limitations of the interrogating sources, detector systems and assay configurations were quantified and complex but rigorous response analysis logic was proposed. Results demonstrate that the delayed gamma-based assay technique has the capability of achieving the desired sensitivity, isotope specificity and accuracy in the verification and accountancy applications. The expected instrument performance was demonstrated for several cases of spent nuclear fuel assemblies calculated with a range of burnups, initial enrichment and cooling times.

V. Mozin, S. Tobin, J.Vujic, A.Hunt, "Delayed Gamma Instrument for Determining Plutonium Mass in Spent Nuclear Fuel," 2010 ANS Annual Meeting, San Diego, CA USA (2010) (LA-UR 10-00561).

V. Mozin, S. Tobin, J.Vujic, "DGSDEF: Discrete Gamma Source DEFinition code", Los Alamos National Laboratory (2010) (LA-CC-10-083).

Passive Gamma measurements of LWR Spent Fuel at ORNL

Passive gamma measurements of LWR spent fuel are currently being conducted at Oak Ridge National Laboratory. The fission fragment gamma peak area ratios are being examined along the length of several PWR fuel rods (including some MOX fuel) followed by confirmatory destructive analysis. A primary goal of the project is to examine possible reasons for a bias in the shipper/receiver difference at the head end of reprocessing facilities. An additional goal is to apply these measurements to the design of a deployable tool to measure passive gamma signatures of spent fuel assemblies in order to validate reactor operator declarations for safeguards. The measurement data, simulations and destructive analysis will be compared so that sources of error can be examined and benchmark data can be cataloged. Figure 2 shows the measured fission product ratios of $^{106}Ru/^{137}Cs$ and $^{134}Cs/^{137}Cs$ along the length of an LWR spent fuel rod.

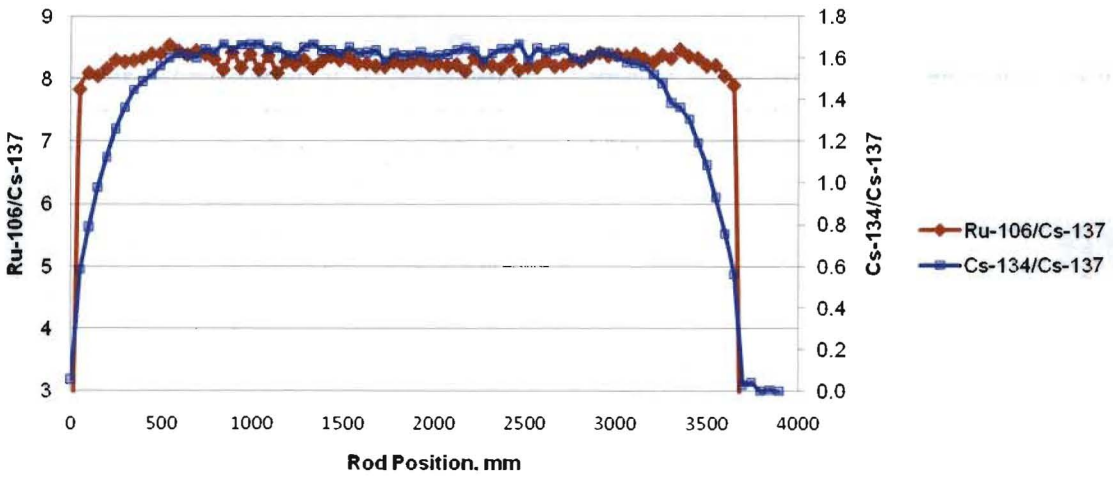

Figure 2. The measured fission product ratios of $^{106}Ru/^{137}Cs$ and $^{134}Cs/^{137}Cs$ along the length of an LWR spent fuel rod.

PNAR-^{3}He Technique

PNAR involves making a measurement with and without a Cd liner on the inner surface of the detector closest to the assembly to form the CD-ratio No-Cd:Cd. A light weight, inexpensive system that uses fission chambers (PNAR-FC) [XXX] that measures gross neutron counts. Here we outline a high efficiency ^{3}He based system (PNAR-^{3}He) that allows Doubles and possibly Triples to also be exploited.

J.L. Conlin and S.J. Tobin, "Determining Fissile Content in PWR Spent Fuel Assemblies Using a Passive Neutron Albedo Reactivity with Fission Chambers Technique," Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, MD (July 11-16, 2010).

The PNAR-^{3}He technique is based on the concept of Passive Neutron Albedo Reactivity (PNAR) and implemented with ^{3}He gas-filled proportional counters for the high efficiency detection of neutrons from temporally correlated events. The design of the PNAR-^{3}He detector hardware is shown in Figure XXXX. PNAR was first proposed by Menlove and Beddingfield in 1997 and applied to neutron multiplicity measurements of uranium fuel rods [XXXX]. PNAR utilizes the self-interrogation of the spent nuclear fuel assembly via reflection of neutrons born in the fuel back in to the assembly. The neutrons originate primarily from spontaneous fission events (e.g. ^{244}Cm) and (α, n) reactions (e.g. oxides) within the fuel itself but are amplified by multiplication. The presence and removal of a Cd liner (~1mm thick) between the reflecting boundary and the assembly provides two measurement conditions with different neutron energy spectra and therefore different interrogating neutron characteristics. In the case with the Cd liner removed, reflected low energy neutrons (thermal neutron albedo) are incident on the fuel assembly and the number of induced fissions, hence neutron multiplication within the fuel, are increased. This amplifies the original spontaneous neutron emission from the fuel. Cd has a high cross-section of absorption for low energy neutrons (< 0.5 eV) therefore the presence of the Cd liner greatly reduces the number of low energy (primarily thermal) neutrons returning to the fuel. PNAR is thus used to assay the fissile content of a spent nuclear fuel assembly by detecting the change in multiplication in the spent fuel assembly between these two measurement conditions using a parameter called the Cd ratio. This is the ratio of the counting rate obtained without Cd in place to the counting rate obtained with Cd present. Improvements in discriminating between fuel assemblies using the Cd ratio are obtained when using doubles counting rates compared with the use of singles counting rates.

The optimum gate width is delayed in time and dependent on the fuel assembly. This has required a new way of thinking.

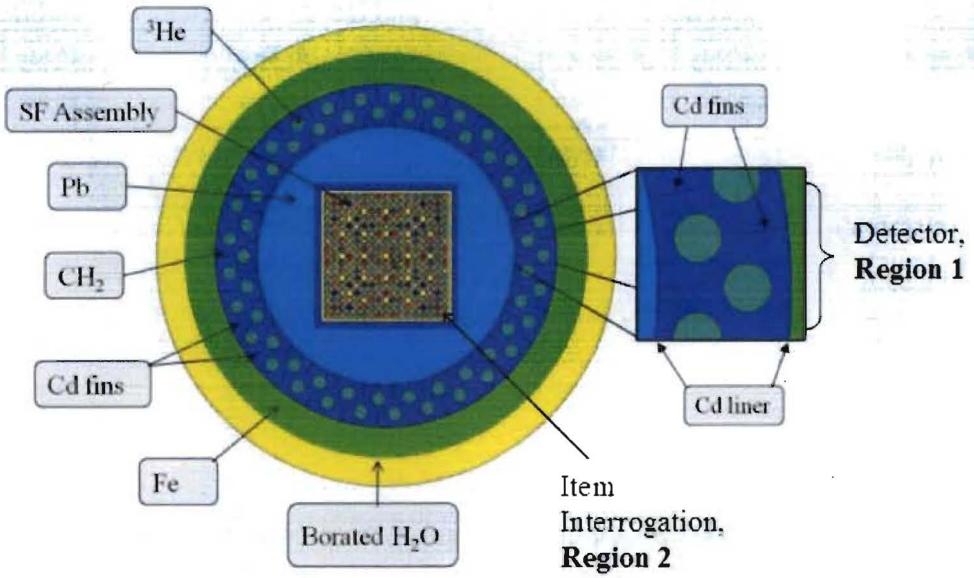

Figure 1. MCNPX Design for PNAR-^{3}He Detector Hardware

[XXX] H.O. Menlove and D. Beddingfield, "Passive Neutron Reactivity Measurement Technique", In Proceedings of the Institute of Nuclear Materials Management Annual Meeting, 1997.

The Differential Die-away Self Interrogation Concept

The DDSI technique is similar to traditional differential die-away analysis, but it does not require a pulsed neutron generator or a pulsed beam accelerator, and it can measure the mass of the spontaneous fission isotope in addition to the fissile mass [1, 2]. The new method uses the spontaneous fission neutrons from ^{244}Cm within the assembly as the "pulsed" neutron source. The time correlated neutrons from the spontaneous fission and the subsequent induced fissions are analyzed as a function of time after the trigger event to determine the spontaneous fission rate and the induced fission rate in the sample. The time separation of the spontaneous fission and induced fission neutrons captured in the detector forms the basis of the technique, and the separate count rates obtained for each lead to the independent measurement of the spontaneous fission mass and fissile mass (^{239}Pu for example). Spontaneous fission mass is determined by multiplicity analysis of the neutrons detected during an early gate, shown in Figure 1, which is opened soon after the initial triggering neutron is detected. Fissile mass is determined from the count rate acquired during the late gate, also shown in Figure 1. Induced fission occurs later in time since the neutrons "interrogating" the fissile mass are actually spontaneous fission neutrons that have been moderated and reflected toward the assembly, i.e. thermal neutron albedo.

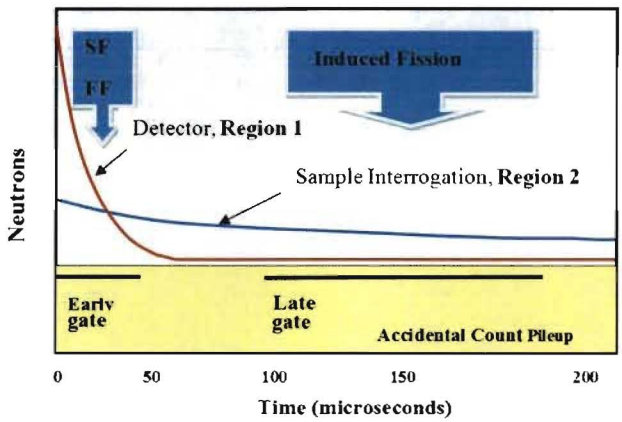

Figure 1: Conceptual neutron capture distributions in the detector from spontaneous fission (SF) and fast fission neutrons (FF) (fast die-away time) and thermal induced fission neutrons (slow die-away time) are used to select early and late gates.

The DDSI detector hardware is the same as that shown for the PNAR-^{3}He system, shown in Figure x. The DDSI detector configuration for spent fuel verification for spent fuel verification has been optimized via Monte Carlo modeling and simulation. The response of the DDSI instrument has been studied for a wide range of PWR assembly cases and two viable fissile ratios have been established. These ratios track with the fissile content present in an assembly when

examined as a function of initial enrichment, burnup and cooling time. A ^{239}Pu effective fissile mass concept formed the basis of translating a measured fissile ratio into a quantitative mass measurement. The fissile ratios were heavily dependent on burnup, cooling time and initial enrichment of the assembly. A multiplication approach has been proposed to deal with the presence of parasitic neutron absorbers which diminish the detected DDSI response.

H.O. Menlove, S.H. Menlove, S.J. Tobin, "Verification of Plutonium content in Spent Fuel Assemblies Using Neutron Self-Interrogation," LA-UR-09-03715, Institute of Nuclear Materials Management 50th Annual Meeting, Tucson, AZ (July 12-16, 2009).

M. A. Schear, H. O. Menlove, S. J. Tobin, S. Y. Lee, L. G. Evans "Fissile Material Measurements using the Differential Die-Away Self-Interrogation Technique," LA-UR-10-04602, Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, MD (July 11-15, 2010).

M. L. Fensin, S. J. Tobin, N. P. Sandoval, S. J. Thompson and M. T. Swinhoe "A Monte Carlo Linked Depletion Spent Fuel Library for Assessing Varied Nondestructive Assay Techniques for Nuclear Safeguards," American Nuclear Society's Advances in Nuclear Fuel Management IV, Hilton Head Island, South Carolina (2009).

Gross Neutron

For commercial light water reactor spent fuel the gross neutron output in a given assay configuration scales with burnup to about the 3rd or 4th power following the build-up of spontaneously fissile higher actinides, especially Cm isotopes. In association with an inventory code prediction and decay correction this provides for a powerful fuel verification. Through the FORK/PYTHON/SMOPY implementations the method is well established for field use [XXX] although opportunity for enhancements and modernization exist. Consequently we shall not discuss gross neutron counting further other than to note that it may be combined with the other neutron instruments discussed.

A. Lebrun, M. Merelli, J-L. Szabo, M. Huver, R. Arlt, and J. Arenas-Carrasco, "SMOPY a New NDA Tool for Safeguards of LEU and MOX Spent Fuel," International Atomic Energy Agency report IAEA-SM-367/14/03 47 (2003).

Neutron Multiplicity Counting

NMC is extensively used for waste and safeguards assay of spontaneously fissile materials [XXXX]. In the case of spent fuel assemblies we can define a ^{244}Cm-effective mass and use a instrument such as PNAR-^{3}He to perform conventional known efficiency analysis of shift-register derived Singles, Doubles and Triples rates. The immediate challenge is one of counting precisions on the Triples rate which is strongly affected by Accidental coincidences at high rates (and for an initial enrichment of 4wt%, 45 GWd/tU, 5 year cooled fuel the count rate is ~6.4MHz, for 5wt%, 60GWd/tU, 5 years cooled fuel the rate is approaching 15 MHz). If we take 1 hour as a reasonable counting time at the pool side precision on the Singles rate is excellent, limited by non-Poisson processes in the electronics due to temperature drifts etc., precision on Doubles is also excellent <~0.2%, but for Triples early indications are that for low BU fuel (one or two reactor cycles) precision is in the percent range while for end-of-life fuel the precision is in the 10-20% range. This is significant because in the present regime the fractional statistical uncertainty on the Multiplication is about 1/3rd that on the Triples rate while the fractional statistical uncertainty on the $^{244}Cm_{eff}$ mass is about three times that on the Triples rate. Thus, non withstanding the biases introduced by the point model equations for interpreting the data Triples counting does not seems attractive for routine assay of end of life fuel. However, since the (α,n) contribution is small and rather uninteresting, and the conventional neutron coincidence counting (NCC) is more robust to violations of the point model assumptions because empirical calibration parameters can be used to compensate, NCC remains candidate for routines (as opposed to special examination) purposes.

N. Ensslin, W.C. Harker, M.S. Krick, D.G. Langner, M.M. Pickrell and J.E. Stewart, "Application guide to neutron multiplicity counting," Los Alamos National Laboratory Report LA-UR-98-4090.

^{252}Cf Interrogation with Prompt Neutron

The CIPN instrument modeled is illustrated in Fig. XXX. Neutrons are detected in ^{235}U fission chambers tolerant of the high gamma radiation fields. A passive (background) is taken followed by an active measurement in which a ^{252}Cf source is moved next to the fuel where it remains stationary for ~100 sec to acquire sufficient precision. The difference between the two count rates is proportional mainly to the thermal multiplication since the measurement is made in water. Fission in ^{235}U, ^{239}Pu and ^{241}Pu dominate but two non-fissile mass contributions need consideration: direct detection of the ^{252}Cf neutrons and fission of ^{238}U; both contribute ~6% ± 2% to the total signal.

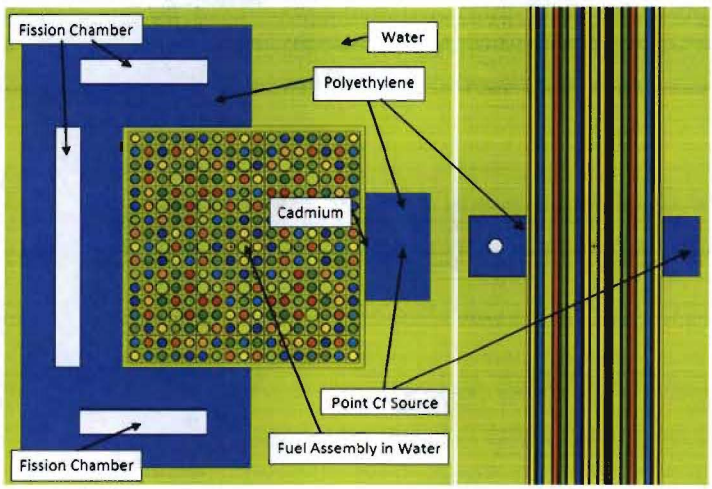

Fig. 1 Plan and vertical cross sections of CIPN (not to scale).

The ^{252}Cf source location was selected to (a) lower the neutron energy in order to minimize fission in ^{238}U and emphasizes fissile isotopes, and (b) provide a large solid angle for the ^{252}Cf neutrons. The arrangement of the fission chambers was selected to achieve nearly uniform count rate per unit mass across the assembly. The thickness of polyethylene was chosen to maximize the count rate.

For convenience the signal is plotted against the effective ^{239}Pu fissile mass defined as a weighted sum of the dominant fissile nuclides ^{235}U, ^{239}Pu, and ^{241}Pu. The CIPN specific weighting factors, which partition the signal, relate the net production of neutrons from fission in ^{235}U and ^{241}Pu respectively including the negative contribution to neutron production by absorption, can be estimated from the fundamental physical processes taking place [XXX].

The calculated behavior is shown in Figure XXX. The data points generally fall into groups of 4 for assemblies that have the same IE and BU but different CT. We see that the count rate trends with effective fissile mass and note structure dependent on BU, IE and CT. In large part this is due to the variation in fission product absorber concentrations. The CT dependence for example in part comes about because ^{155}Eu decays to ^{155}Gd which have a far greater absorption cross section while the decay of ^{241}Pu into ^{241}Am amounts to a loss of fissile and a gain in absorber. The challenge is to parameterize the structure in terms of the gross variables so that the necessary corrections can be applied with minimal prior knowledge.

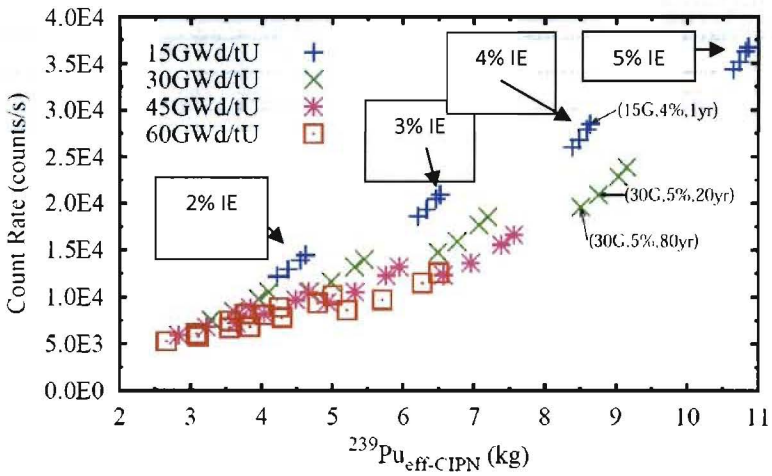

Fig. 2. CIPN count rate vs. $^{239}Pu_{eff\text{-}CIPN}$ for the 64 assemblies in the spent fuel library for a ^{252}Cf source of 2 x 10^8 n/s. The BU, IE and CT for 3 assemblies are individually labeled to give the CT structure.

Ref

J. Hu, S. J. Tobin, H. O. Menlove, "Determining Plutonium Mass in Spent Fuel Using ^{252}Cf Interrogation with Prompt Neutron Detection," Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, MD (July 11-16, 2010).

Self-Interrogation Neutron Resonance Densitometry

The use of SINRD to quantify the fissile content in LWR spent fuel is a promising technique for the improvement of nuclear safeguards and material accountability measurements. The idea is that by measuring the relative neutron flux changes in several narrow energy ranges, the mass of fissile isotopes such as ^{235}U and ^{239}Pu can be determined. The sensitivity of SINRD is based on using the same fissile materials in the fission chambers as are present in the fuel because the effect of resonance absorption lines in the transmitted flux is amplified by the corresponding (n,f) reaction peaks in fission chamber. Ratios of different fission chambers are used to reduce the sensitivity of the measurements to extraneous material present in fuel. This also reduces the number of unknowns because the neutron source strength and detector-fuel assembly coupling cancels in the ratio. In addition to bare FCs one can also used FCs wrapped with carefully chosen absorbers to alter the response. For instance a ^{235}U FC covered with 3 mm of Cd primarily detects neutrons with energies above ~2 eV, while a ^{235}U FC covered with 0.1 mm of Gd detects neutrons with energies above ~0.2 eV. The difference between these two measurements gives the flux between ~0.2 and ~2 eV where ^{235}U has two resonant peaks. A "fast flux monitor" used for normalization can be constructed by surrounding a ^{235}U FC in B_4C. The SINRD method provides a number of potential improvements over current IAEA verification methods. These improvements include:

1. SINRD provides absolute measurements of burnup independent of the operator's declaration and is insensitive to the concentration of boron in the water and the initial enrichment of the fuel. Thus, SINRD can be used at multiple spent fuel storage facilities.
2. The calibration of SINRD at one reactor facility carries over to reactor sites in different countries because it uses the ratio of fission chambers that are not facility dependent.
3. SINRD can distinguish fresh and 1-cycle spent MOX fuel from 3- and 4-cycles spent LEU fuel without using reactor burnup codes.

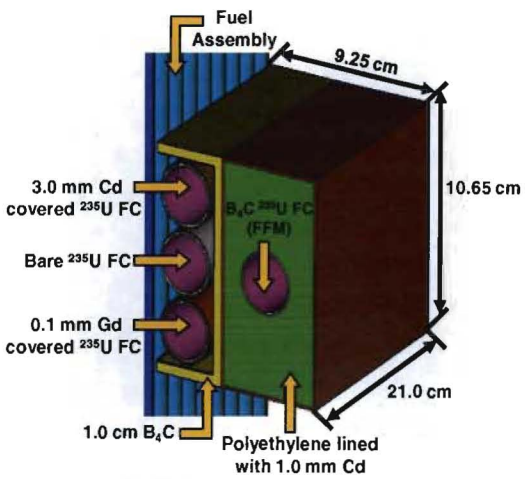

Fig. XXX. Schematic of a ^{235}U FC SINRD detector.

A.M. LaFleur, H.O. Menlove, W.S. Charlton and M.T. Swinhoe, "Development of self-interrogation neutron resonance densitometry to measure the fissile content in spent fuel," Los Alamos National Laboratory report LA-UR-09-08178 (Jan., 2010).

A.M. LaFleur, W.S. Charlton, H.O. Menlove, M.T. Swinhoe, S.Y. Lee, and S. J. Tobin, "Experimental Benchmark of MCNPX Calculations Against Self-interrogation Neutron Resonance Densitometry (SINRD) Fresh Fuel Measurements," Institute of Nuclear Materials Management 51st Annual Meeting, Baltimore, MD (July 11-16, 2010).

X-Ray Fluorescence Measurements of LWR Spent Fuel at ORNL

The direct measurement of x-ray lines from passively induced x-ray fluorescence (XRF) in LWR spent fuel has been achieved for the first time at Oak Ridge National Laboratory (ORNL). The results of initial measurements show that the ratio of the Pu to U x-ray peak areas have a linear correlation to the Pu/U content at the outer edge of the fuel pin as determined by MCNPX simulations completed at Texas A & M University. Results of destructive analysis are pending and will be used to determine the correlation between the measured Pu/U ratio to the total Pu/U content in the fuel. Figure 1 shows the measured 103.7 keV Pu x-ray peak on the left and the 105 keV ^{155}Eu peak on the right.

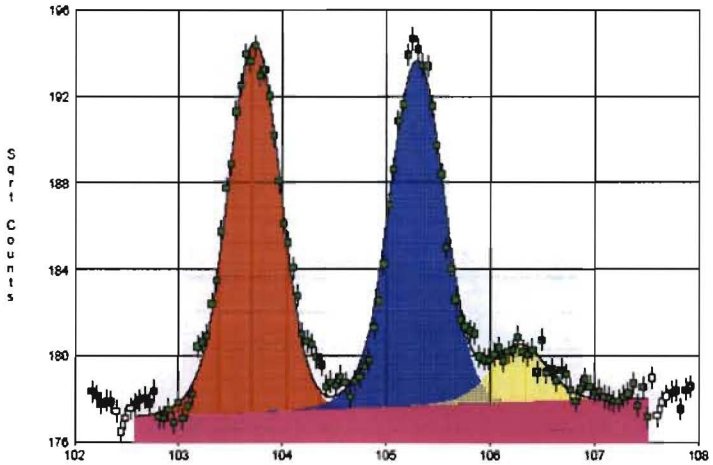

Figure 1. Example of the measured x-ray peak for Plutonium. The 103.7 keV Pu x-ray peak is on the left and the 105 keV ^{155}Eu peak is on the right.

Lead Slowing Down Spectrometer

LSDS is an active neutron interrogation technique which has not been considered as a NDA instrument on this scale before. In concept the assembly would be placed in a large lead cavity. A pulsed fast-neutron source, such as an electron linear accelerator, would fire. The neutrons are quickly slowed by inelastic scattering to where elastic scattering is responsible for moderation. The energy loss per scatter is slight so the energy spectrum persists and cools gradually. Faster neutrons have their next collision earlier and so the burst tends to maintain a consistent relative energy spread as the mean energy falls. As the interrogating neutrons sweep across the resonance structure of the fissile nuclides present induced fission neutrons can be detected. The characteristic time patterns recorded in isotopic fission chambers allow the fissile nuclides of interest, such as ^{235}U, ^{239}Pu and ^{241}Pu to be unfolded from basis vectors. Resonant absorbers such as ^{240}Pu can be extracted by their influence on the self-shielding factor. The presence of hydrogen, even at the level create in the cladding, perturbs the slowing down process, as do the light elements in the fuel. Measurements must be performed in air.

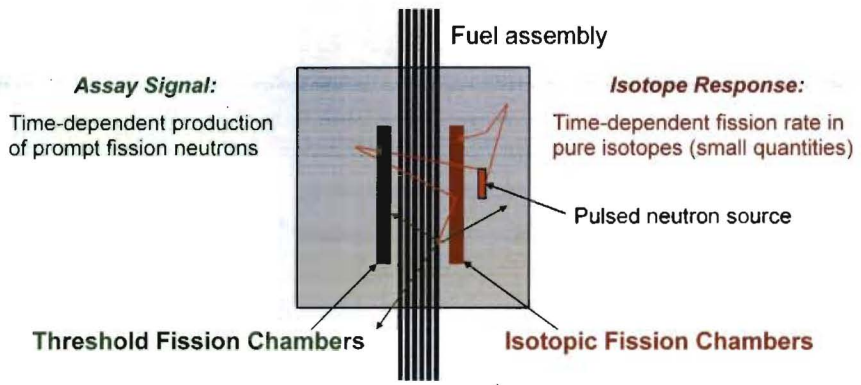

Fig. XXX. Schematic of a nominal lead-slowing-down spectrometer for spent nuclear fuel assay.

The LSDS instrument appears to have good spatial uniformity which together with isotope specificity makes it sensitive to pin diversions. However a large volume of high-purity lead (likely greater than 25 tons), an intense pulsed neutron source, and a suitable assay facility are needed likely limiting the approach to for example the head-end of a reprocessing plant, mixed-oxide fuel fabrication plants (particularly for those fuels where gamma-ray and neutron signatures from recycled actinides overwhelm Pu emissions), and interim or long-term dry storage facilities. Work is progressing on temporal analysis algorithms to deal with the non-linear response to the cocktail of nuclides present in spent fuel and on ways to represent spectral perturbation and self-shielding. Analytical models have proven useful since direct modeling challenges Monte Carlo techniques.

L. E. Smith et al., "Advancements in Time-Spectra Analysis Method for Lead Slowing-Down Spectrometry," Proceedings of the 2010 ANS annual meeting, San Diego, California. (2010).

Neutron resonance transmission analysis

NRTA is an isotopic-assay technique suitable for assaying commercial nuclear reactor spent fuel. The NRTA technique uses low-energy neutrons in the 0.1-eV to 40-eV energy range as a probing radiation and measures the degree to which these are attenuated as they traverse a spent fuel assembly. The NRTA concept is not new; indeed, it is one of the older demonstrated nondestructive fuel assay methods for plutonium analysis. The NRTA concept is based on solid theoretical principles has been demonstrated experimentally at the bench scale using commercial spent fuel; NRTA has achieved a plutonium assay measurement precision of 2-4% in ad-hoc testing [1-4]. It originated with research at the National Bureau of Standards (NBS) in the early 1970s, research on the use of NRTA for spent fuel analysis continued at the NBS into the mid-1980s. The low-energy neutrons needed for NRTA measurements are produced using a pulsed accelerator; for example, a 10-MeV electron accelerator with a beryllium photoneutron converter and neutron moderator such as deuterated water. The entire assembly is operated in a time-of-flight arrangement. Neutrons travel from the photoneutron production area down a drift tube of 2 to 4 meters length in a pencil beam or fan beam collimator geometry. A spent fuel assembly is placed at the end of this drift tube; on the opposite side of the fuel is a second drift tube and collimator of comparable length, with a 1- or 2-dimensional neutron detector at the far end. A schematic representation of the NRTA layout is shown in Figure 3.

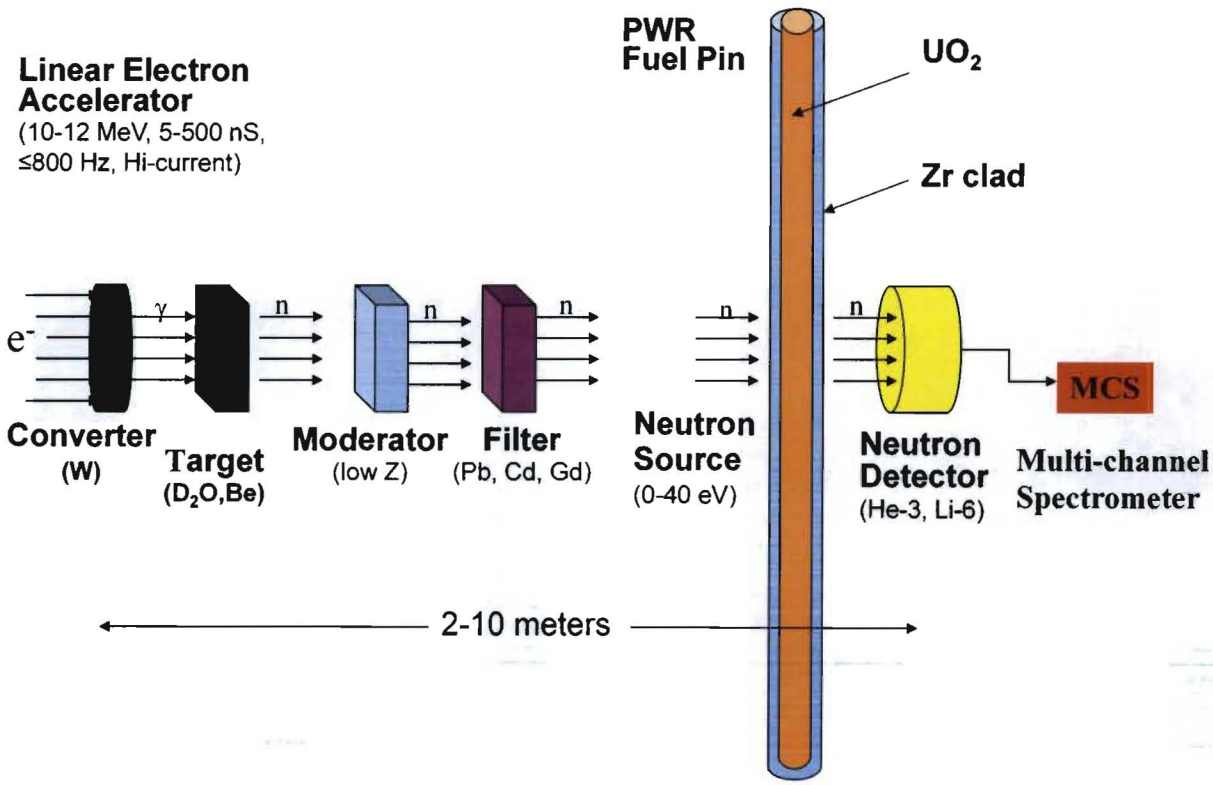

Figure 3 Basic NRTA concept and component layout.

On-resonance neutrons are preferentially scattered out of the neutron beam as they traverse the spent fuel, in proportion to the abundance of each isotope in the fuel and the resonant neutron attenuation cross-sections of those isotopes. Faster neutrons reach the fuel before slower neutrons; timing data is collected from the detector where neutron counts are recorded versus the time following each accelerator pulse. Early event times correspond to higher-energy neutrons while later events correspond to lower-energy neutrons. Higher event rates indicate the lack of attenuating materials in a particular energy range while lower event rates indicate the presence of materials with higher (resonant) absorption characteristics at a particular energy. The energy dependent profile of neutron absorption through the fuel is correlated with the presence of individual isotopes with neutron absorption resonances in the beam path.

The NRTA neutron energy range is at the bottom end of the actinide resonance range, where most actinides have at least one or more resonances. For the actinides found in spent fuel the resonances are typically large in magnitude, narrow in breadth, and fortuitously well-separated, resulting in distinctive resonance transmission spectra. NRTA is capable of directly assaying ^{235}U, ^{236}U, ^{238}U, ^{239}Pu, ^{240}Pu, ^{241}Pu, and ^{242}Pu in spent fuel. It is also capable of directly assaying the fission products ^{145}Nd, ^{133}Cs, ^{99}Tc, ^{152}Sm, ^{131}Xe, and ^{103}Rh. The optimal approach for NRTA measurements would be to use a fan-beam geometry and to inspect fuel assemblies length-wise in 5-10 cm increments. Practical NRTA beams will only be capable of performing complete assays through a stacked arrays of 8 to 12 fuel pins. Because of this, multiple angle views will be required to completely scan each assembly.

Research is underway at Idaho National Laboratory to study the applicability of the NRTA technique for meeting the spent fuel plutonium measurement goals of the NGSI program. This work is focused on using advanced simulation and modeling a) to benchmark the experimental results presented in the literature from previous NRTA measurements of spent fuel pins and b) to conduct parametric studies to assess the capabilities of NRTA for assaying complete spent fuel assemblies. Strengths of the NRTA technique are that it directly assays plutonium without reliance on correlations, that it is extremely sensitive for the detection of fuel pin diversion and is resistant to spoofing, and that it is capable of providing additional fuel assay information including fuel burn-up. Weaknesses of the NRTA technique in comparison with other types of spent fuel safeguards measurements are its complexity and the large infrastructure requirements needed for NRTA measurements.

H. G. Priesmeyer and U. Harz "Isotopic Assay in Irradiated Fuel by Neutron Resonance Analysis," Atomkernenergie (ATKE) 25 (1975) 109-113.

R. A. Schrack et al. "Resonance Neutron Radiography using an Electron Linac," IEEE Trans. Nucl. Sci. 28 (1981) 1640-1643.

C. D. Bowman, C.D., et al., "Neutron Resonance Transmission Analysis of Reactor Spent Fuel Assemblies," *Neutron Radiography*, Barton, J. P. and von der Hardt, P., eds., ECSC, EEC, EAEC, Brussels, Belgium and Luxembourg (1983) 503-511.

J. W. Behrens, R. G. Johnson and R. A. Schrack "Neutron Resonance Transmission Analysis of Reactor Fuel Samples," Nucl. Tech. 67 (1984) 162-168.

Nuclear Resonance Fluorescence

NRF is the process by which a nucleus is excited to a nuclear state by the absorption of a photon, and then subsequently de-excites to the ground state by the emission of one or more gamma-rays. The energies of the photons that induced the initial excitation, and the energy of the re-emitted photons are characteristic of the specific state that underwent NRF and therefore characteristic of the isotope. For the assay fissile materials photons between about 1.5 and 4 MeV are most useful. NRF has not previously been studied for SNF assay. Because NRF generates isotope-specific signals and the promise and appeal of the technique is its potential to directly measure the amount of a specific isotope in SNF.

Both, the elastic and inelastic scattering of the interrogating photons generate background that is largely forward directed. The characteristic NRF gamma-rays emitted must be therefore detected at backwards angles where the backgrounds are lowest. This is called the backscatter geometry. An additional important contribution to the background is the gamma radiation from the radioactive decay of the fission products in SNF. The great challenge is to achieve the required measurement sensitivity for quantifying the mass of Pu isotopes in SNF assemblies since ^{239}Pu concentrations are low, the cross-sections are very narrow so that Doppler broadening an other subtle physical affects are influential, and obtainin an intense radiation source with desirable properties is non trivial.

The alternative is to use the transmission method schematically illustrated in Figure XXX. Transmission assay uses a detection systems down-stream of the assay target to measure the excess attenuation of resonant-energy photons in the assembly. The detection system consists of a thin sheet of material containing the isotope of interest called the witness foil and a detector array that measures the NRF gamma-rays coming off it. Given the low concentrations of the Pu isotopes in SNF and the small integrated nuclear resonance cross sections, the main challenge for both methods in achieving the goal of 1% measurement accuracy lies in the accruing of sufficient counting statistics in an acceptable measurement time.

To quantify the potential of these techniques for the direct measurement ^{239}Pu an analytical model was developed. Extensive MCNPX modeling was also performed to calculate and understand the scattered, non-resonant background, the notch refilling in transmission measurements, and to fully simulate NRF measurements on SNF assemblies. It was found that MCNPX underestimated the non-resonant elastic scattering at backwards angles by multiple orders of magnitude mainly due to a flawed treatment of Rayleigh scattering in the code. An improved treatment gave much better agreement with laboratory experiments.

The backscattering method has three main limitations: a low signal to background ratio for the small Pu concentrations in SNF, a high background from the radioactive decay of the fission products in the spent fuel, and a strong dependence on depth of the intensity of the NRF signal. These difficulties render this approach not viable for the accurate measurement of low concentration of Pu isotopes in SNF with bremsstrahlung sources with an end-point energy just above the resonance of interest.

The transmission method provides two important advantages: first, the detectors can easily be shielded from gamma-rays emitted from the fission products in the SNF, and second, the measurement sensitivity is not depth dependent. In this method the areal density of the Pu isotope is derived from the decrease of the NRF peaks in the measured spectrum. This decrease is on the order of 0.5% for the known ^{239}Pu resonances implying very good counting statistics are needed to determine this quantity with 1% accuracy. Due to the high flux of mostly lower energy, scattered non-resonant photons associated with the bremsstrahlung beam the count rate limited detectors need to be operated behind thick filters, which reduce the already low NRF count rate further. Sufficiently precise measurements of Pu isotope concentrations in SNF would require 10's to 100's of hours, a very intense bremsstrahlung source, and a very large detector array.

It can be concluded, based on this study, that NRF-based methods for practical, direct, ^{239}Pu assay in SNF assemblies cannot presently be realized using bremmstrahlung sources and high resolution gamma-ray spectrometers. Measurements with the required sensitivity and accuracy most likely require quasi-monoenergetic photon sources with intensities that are at least two orders of magnitude higher than those currently being designed or proposed. Such advancements may become available in the future, but substantial progress in electron accelerator and laser technologies is still needed.

Quasi-monoenergetic photon sources such could potentially provide much enhanced capabilities and result in significantly different conclusions. Demonstration sources that produce low divergence, narrow beams of 2-2.5 MeV photons with a 1-10 keV energy spread are currently being constructed and more intense sources are being proposed. The times needed to reach 1% counting precision in a transmission measurement on an SNF assembly with 0.4% ^{239}Pu content for a range of source parameters and detector options. As an example, for a photon sources with a 1 keV energy spread, an intensity of 6×10^8 ph/eV/s, and operating continuously or at MHz pulse rates the measurement time would be 7 hours assuming an array of 100 $LaBr_3$ detectors (100% relative efficiency).

Threshold detectors, such as Cerenkov detectors, that can integrate the signal instead of detecting single photons are not rate limited and thus would enable measurements with pulsed beams.

Measurement times would be limited only by the intensity of the photon source. Threshold detectors would also make backscatter measurements with very high intensity pulsed photon sources possible. The detectors could be gated and thus be insensitive in between short, sub-nanosecond pulses. This would reduce the background from the radioactivity of the SNF seen by the detectors by many orders of magnitude.

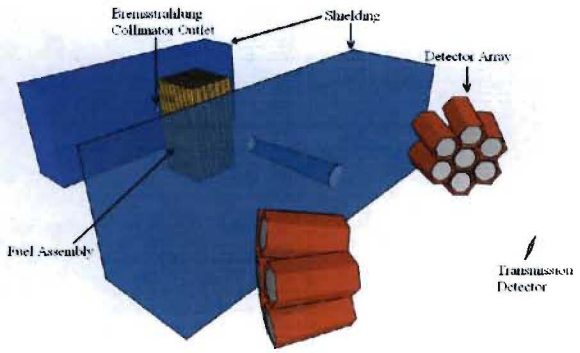

Figure XXXX Schematic description of a geometry used for a transmission assay measurement

Conclusion

We have briefly outlined the NDA methods being evaluated as part of the NGSI multi-lab/university collaboration to quantify the Pu mass in, and detect the diversion of pins from, spent nuclear fuel assemblies. This work is on-going. Some techniques are familiar but have benefited from renewed interest which has improved nuclear data and simulation. Other methods are novel requiring basic scientific development. Individual methods are complementary. Future work will consider how to integrate a few of the most promising techniques to create a practical and robust means to assay spent fuel assemblies for Pu. This will involve advanced modeling work and full scale proving experimental work in collaboration with international partners.

Acknowledgement

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